Comment on "Structure factors of harmonic and anharmonic Fibonacci chains by molecular dynamics simulations"

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Recently, Engel et al. discussed phonon broadening as observed in 3D quasicrystals on the basis of calculations on the Fibonacci chain. We show that the paper contains several statements and assumptions that are contradicted by factual truth.

Recently Engel et al. [1] provided a discussion of the phonon dynamics of the Fibonacci chain. Their aim was to explain the broadening of phonons observed in experiments. This comes just after the rejection of a Comment of mine [2] on reference [3] in which I developed a first-principles discussion, that is both rigorous and pertinent, and - in contrast with reference [1] - does not introduce unnecessary assumptions. To reject my Comment it was argued that a study of one-dimensional QCs would be futile and irrelevant for real 3D QCs, while a co-author of reference [3] had very obviously claimed the opposite on another occasion [4].

There are other mechanisms that can explain phonon broadening than the one proposed by the authors, such that the conclusions the authors try to draw from their models are not compelling. Moreover the models introduced by Engel et al. are not realistic. The phonon-like mechanism that is proposed for the jumps, and illustrated in Figure 1, is in contradiction with experimental data for two reasons. (1) Phonon broadening is clearly observed at room temperature, while atomic jumps are not. Moreover there are no data that warrant the assumption that there would exist some anharmonicity at low temperatures. (2) This assumption is perhaps introduced on the basis of a conviction that the atomic jumps could not be explained otherwise. But already from the text book case of self-diffusion in normal periodic solids, it should be clear that there exist other possibilities: In the case of self-diffusion atomic jumps are not a consequence of anharmonic potentials but of the presence of vacancies. Experimental data show that atomic jumps in quasicrystals are *not thermally activated* as postulated by the authors in their model, *but assisted*. Indeed, it is not the *width* of the quasielastic line that follows an Arrhenius behaviour in the neutron data as it should be with a thermally activated process but the *intensity*. The assisting mechanisms have never been identified with certainty and they could be multiple. One possibility could be the presence of vacancies that make the environment more loose and thereby lower the barriers against the jumps, but such a scenario is not cogent. In certain cases (e.g. AlCuFe) the value of the assistance energy determined is so high (0.6 eV) that this alone already precludes any identification with some potential barrier ΔE in a phonon-driven mechanism according to Figure 1, because the resulting Boltzmann factors are too small even close to the melting point.

Two claims about the incoherent structure factor are wrong:

(1) There can be no Lorentzian with a width $\Gamma \propto Q^2$ without long-range diffusion. Hence the result for the harmonic periodic chain is unphysical and an artifact of the treatment. In fact, following the logic of the authors, real tridimensional harmonic periodic monoatomic crystals should also exhibit Lorentzians, which according to real experimental data they clearly do not. The HPC calculation should not yield any physically meaningful Lorentzian. The fact that such a Lorentzian is nevertheless present even in the HPC, shows that all conclusions drawn from the presence of Lorentzians in other models are biased because these Lorentzians have at least a component that is unphysical.

(2) The general statement that randomizing the Fibonacci chain due to the presence of atomic jumps would not change the incoherent structure factor because the jumps are local is just plain wrong. If only things were that easy! A clear counter example that shows how the argument that only the local environment would define the signal leads to completely wrong conclusions is the case of an atom that makes first-neighbour jumps with relaxation time τ between N sites equally spaced on a straight line. The solution of this problem depends explicitly on the value of N. The fact that a single jump is local cannot be used to justify simplistic folk lore about the incoherent structure factor. The only reliable way to make rigorous statements about the incoherent structure factor is solving the problem, e.g. in configuration space as discussed in reference [5], where it is shown that also in incoherent scattering the configurations have to be taken into account and that the number of relevant configurations is even higher for incoherent scattering than for coherent scattering. Furthermore the statement introduces an arbitrary if not meaningless distinction. The presence of atomic jumps is not possible without randomization. And heavy randomization introduces a possibility for lang-range diffusion that is absent from a model with sufficiently mild randomization.[6]

It is unappropriate that the autors use a completely unrelated paper about phonon dynamics as a pretext to comment on an issue of terminology and/or presence of atomic jumps in quasicrystals. They totally misrepresent this issue, following in this respect reference [7]. These authors [7] deliberately misinterpreted a statement on my behalf

that fast atomic jumps are typical of the local quasicrystalline structure (in the sense that they are a consequence of the local quasicrystalline structure ($A \Rightarrow B$)) as a statement that fast atomic jumps would only occur in quasicrystals ($B \Rightarrow A$). This inversion of the logical implication was then used to argue that phason jumps are nothing special, because fast atomic jumps can be found in some crystals as well. This denegration methodology is analogous to claiming that the platypus is a not very special animal since birds are laying eggs as well. Moreover the choice of the sample that should serve as a "counter-example" was deliberately biased in that it has an exceptionally high concentration of structural vacancies.

The importance of the work on phason dynamics in QCs resides in the fact that it showed that phasons in quasicrystals are atomic jumps (and not modes). That was a very important clarification, especially when we see that even 15 years later certain people still refuse to accept it, as their recent papers clearly show. That fast atomic jumps also occur in some crystals cannot make away with the fact that they correspond to the concept of phasons in quasicrystals while they do not in crystals. And claiming that atomic jumps that are this fast are nothing special is just dishonest. The same can be said about the conduct of Peter Adams with respect to reference [2].

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